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ABSTRACT

Coincidence counting techniques were used in the determination of the half-life of Hf-178m. The experimental sample, essentially a mixture of Hf-177 and Hf-178, was irradiated with thermal neutrons to produce Hf-178m and Hf-179m isomers. The isomeric transition of Hf-178m is followed by a cascade of gammas (.427, .326, .214, and .093 mev). The isomeric transition of Hf-179m (approximately 18-second half-life) is followed by a .215 mev gamma. The decay data of Hf-178m was effectively isolated from that of Hf-179m by counting coincidences between any two of three specific Hf-178m gammas (.427, .326, and .214 mev). After being corrected for chance coincidence counts, this counting data was processed by a computer program (FRANTIC). By using the least-squares techniques, the FRANTIC program fitted a calculated exponential decay curve to the counting data from a single experiment and also computed the half-life for that data. The average half-life of Hf-178m, derived from seven decay experiments, had a value of 3.94 seconds and an uncertainty of 0.5%. The additional uncertainties due to gain instability and background count-rate were estimated. The half-life of Hf-178m was found to be 3.94 ± 0.04 seconds, all uncertainties included.

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1. Introduction.

Two (n,γ) reactions leading to isomeric states take place when a Hafnium sample containing Hf-177 and Hf-178 is irradiated with thermal neutrons. The thermal neutron activation cross sections of Hf-177 and Hf-178 which lead to isomeric states are 1.4 and 50 barns respectively. The first reaction, Hf-177 plus thermal neutron going to Hf-178m, produces an isomer whose half-life is of the order of 4 seconds. The second reaction, Hf-178 plus thermal neutron going to Hf-179m, produces an isomer whose half-life is of the order of 18 seconds.

The half-life of Hf-179m has recently been determined to a degree of accuracy deemed to be better than 1% (private communication from Rodeback, G. W.); therefore, it will not be pursued in the present investigation. The half-life, in seconds, of Hf-178m has previously been reported as 4.8, 3.5, 4.3, 3.9, and 3.8 by Felber, F. F. et al, Campbell, E. C. et al, Alexander, K. F. et al [3], Griggs, J. C. et al [2], and Button, E. J. et al [1] respectively. The adopted value reported by the National Research Council [3] is 4 seconds. The objective of the present investigation is to more accurately determine the half-life of Hf-178m by employing coincidence counting techniques. A counting system using these techniques can effectively isolate the Hf-178m counts from the combined counts due to both Hf-179m and Hf-178m which are decaying in the activated sample.

The coincidence method measures the simultaneous (within a resolving time t) emission of two types of radiation from the same

nucleus. To accomplish this, a gamma counting system (Figure 5) which includes two detectors and a coincidence module is required. If a pulse from detector number one arrives at the coincidence module within a resolving time t of the arrival of a pulse from detector number two then the coincidence module registers a coincidence count. For purposes of gamma-gamma coincidence counting, the types of radiation refer to the energy ranges of each of the gamma radiations. By placing a single channel pulse height analyzer in the circuit of each detector, the energy (pulse height) range of pulses admitted to the coincidence module from that detector can be controlled.

It is possible for radiations from two different nuclei to impinge on the detectors so closely in time that a false coincidence count results. The contribution to the coincidence rate by such chance counts is called the chance coincidence counting rate. This chance rate is given by the product: (2) (resolving time t) (singles rate in detector channel number one) (singles rate in detector channel number). The true coincidence counting rate is the measured rate less background and chance rate.

In this experiment the energy ranges of the gammas for coincidences were selected to eliminate true coincidences due to Hf-179m decays. For detector channel number one the energy range included all gammas above .15 mev. For detector channel number two the energy range included all gammas in the interval .29+ mev to .48+ mev. This selection thus limited true coincidences to those between the Hf-178m gammas (Figure 1) of energies .427, .326, and .214 mev. A single

.427 mev gamma in detector channel number two could be coincident with either a .326 or a .214 mev gamma in detector channel number one. A single .326 mev gamma in detector channel number two could be coincident with a .214 or a .427 mev gamma in detector channel number one.

The source of thermal neutrons was the A G N - 201 reactor. The thermal neutron activation of the Hafnium sample was effected by a penumatic transfer system. By activating the firing switch, the activated sample (enclosed in a balsa projectile) was propelled,by compressed air,from the reactor core to the site of the two detectors in about one-tenth second. Gamma radiations (Figure 1) from Hf-178m and Hf-179m impinged on both detectors (Figure 6). For coincidence runs (Figure 5), pulses from each detector (NaI(Tl) crystal and photomultiplier tube) passed through a preamplifier and amplifier and into a single channel pulse height analyzer. The analyzer in the circuit of detector number one was adjusted to pass to the coincidence module only those pulses whose energy (pulse height) was above .15 mev. The analyzer in the circuit of detector number two was adjusted to pass only pulses in the range .29+ to .48+ mev. The coincidences sensed by the coincidence module were registered on a scaler. This scaler was modified so that each of its output pulses represented a scaled number of counts. These scaled count pulses, as well as accurate one-second timing pulses, were placed on chart paper by a chart recorder. Additionally, the recorder placed event marks on the chart

margin which indicated the time duration of firing-switch-actuation and sample irradiation.

The data from the recorder charts was first processed by the REDUCTION computer program. Its output, corrected for chance coincidence counting rate, provided the input for the FRANTIC computer program. The FRANTIC program, by performing a least squares analysis on the input data, gave the half-life of Hf-178m and its sigma (estimator of standard deviation).

2. Results.

FRANTIC Output

Run	Half-life (seconds)	Estimator of Standard Deviation
1	3.922	.036
2	3.986	.043
3	3.981	.072
4	3.833	.028
5	3.957	.043
6	3.945	.034
7	3.965	.048
Average	3.94	.02

Discussion

The time data on one side of the chart paper had to be transposed onto the other side where the count data was recorded. To accomplish this, measurements to a common transverse grid line were made to

the estimated nearest hundredth of a centimeter. The resulting uncertainty in the determination of the timing interval was 1% or less. This 1% uncertainty was an input to FRANTIC and hence its effect is reflected in the statistical uncertainty of the computed half-life.

A principal uncertainty in the computed half-life was due to possible gain shifts in the detector channels. Based on the performance of both detectors, noted during this experiment, and the findings of Rodeback and Button et al (see Gain Shift, Section 7), it was concluded that .4% is a reasonable estimate of the above uncertainty.

A lesser, but still important, uncertainty in the half-life determination is due to the background input provided to FRANTIC. Since the background was a fixed input, FRANTIC considered its standard deviation to be zero. By providing FRANTIC with several assumed backgrounds and analyzing the resulting half-lives, the error in half-life due to background uncertainty was estimated to be .1% or less (See Effect of Background Uncertainty on Half-life, Section 7).

The uncertainty in the resolving time of the coincidence module had a negligible effect on the computed half-life (See Effect of Resolving Time Uncertainty on Half-life, Section 7).

A study was made to ascertain whether a true coincidence count could originate from a Hf-179m nucleus. A decaying Hf-179m nucleus has only two cascading gammas (Figure 1) and both them must pile-up in detector channel number two in order to get a pulse into the coincidence module. Hence there is no remaining available gamma from

that nucleus to impinge on detector number one and therefore no true coincidence count can be registered. This was verified by an analysis of a decay curve (run number seven) plotted on semilog graph paper (See Effect of 18 second Hf-179m Component on Half-life, Section 7).

Summary of Uncertainties and
Statement of Half-life of Hf-178m

Uncertainty		Percent
Statistical		0.5
Gain-shift		0.4
Background		0.1
	Total	1.0

Half-life of Hf-178m: 3.94 +.04 seconds

3. Equipment.

Pneumatic Transfer System

Each sample to be irradiated was enclosed in a gelatin capsule (with polyethylene filler) which in turn was inserted into a balsa projectile 1-3/4 inches long and approximately .775 inches in diameter. The projectile was placed in the projectile barrel of the pneumatic transfer system. The system permitted impressing pressure-vacuum differentials on either end of the projectile. Thus the projectile could be rapidly transferred back and forth between counting site and the reactor core. In practice the projectile was pneumatically inserted into the core for time controlled irradiation and then transferred back

to the counting position. Approximately one-tenth second is required for a one-way trip.

Hafnium Sample

The Hafnium sample was prepared by the Oak Ridge National Laboratories. Their assay data was as follows:

Isotope	Atomic Percent	Precision
174	less than .05	---
176	1.15	± 0.05
177	84.52	± 0.1
178	9.2	± 0.05
179	2.22	± 0.05
180	2.91	± 0.05

NaI(Tl) Detectors

The detector designated as number two (Figures 2, 4, and 5) consisted of a type 12A12 Harshaw NaI(Tl) crystal and a Model 9578 Electra Megadyne Incorporated photomultiplier tube. The excellent stability of this detector was reported by E. J. Button et al. [1] The high voltage input was 900 volts

The detector designated as number one (Figures 2, 4, and 5) consisted of a type 12S12 Harshaw NaI(Tl) crystal and a DuMont photomultiplier tube. The high voltage input was 900 volts.

Preamplifiers

A Hamner NB-11 Preamplifier was connected to the output of each photomultiplier tube. The NB-11 has a rated gain stability of .1% per volt change of line voltage. It has a differential linearity of ±1% or better and can accomodate a counting rate of 150,000 counts per second for .1% gain shift.

Amplifiers

Pulses from each preamplifier were routed over approximately 22 feet of RG/58U shielded cable to a variable gain Hamner NA-12D Double Delay Line Linear Amplifier. The amplifier was operated in the double delay mode, a design feature which at high counting rates preserves the linear relation between energy and pulse height. The gain stability is rated at 0.1% per degree centigrade, 0.02% or better per volt of line voltage variation and ±0.1% gain shift with a maximum count rate of 100,000 counts per second.

Single Channel Pulse Height Analyzers

Two Hamner NC-14 Low Jitter Pulse Height Analyzers were used (Figures 4 and 5) for data collection. The NC-14 has the designed capability of selecting precise energy intervals in either the differential or integral modes. In the differential mode its output consists only of pulses falling in the interval E to $E+\Delta E$; in the integral mode its output includes all pulses of pulse height (energy) which are equal to

or greater than the E setting. Furthermore, the analyzer was operated in the crossover triggering mode, a design feature which preserves the time information carried by the input pulses. The output may be put directly into fast coincidence with resolving times in the 2t = 10 to 30 nanoseconds region.

An auxiliary scaler was available to either analyzer and so modified that each output pulse was appropriately shaped for input to the chart recorder. Each scaler was further modified so that each output pulse could represent either 10, 100, 1000, or 10,000 input pulses. Any initially-selected scale factor could be changed easily during runs by means of a four position switch.

The NC-14 has the following specifications:

E:	.3 to 10 volts by 10 turn helipot; stability of 2 mv/ °C or better; linearity of $\pm$.25% of gain setting or better.
ΔE:	0 to 10 v by 10 turn helipot; window width stability of .5mv/ °C or better.
Pulse pair resolution:	1 microsecond.
Count rate dependence:	no measurable shift at E or ΔE thresholds at 500,000 cps.
Coincidence outputs:	350 nanosecond pulse width; rise time less than 15 nanoseconds.

Coincidence Module and Decade Scaler

A Hamner NL-16 Fast Ramp Coincidence Module was used during coincidence data collection runs. It was designed for experiments requiring fast coincidence logic in the 2t = 10 to 30 nanoseconds resolving range. In these experiments it was employed with a helipot dial setting of 5.00 which corresponds to a resolving time t=75 nanoseconds.

Coincidence pulse outputs were routed into a Hamner N-240 Decade Scaler. This scaler was modified in exactly the same manner as the one discussed above. The coincidence module and this scaler will hereafter be referred to as the coincidence module-scaler linkup.

Crystal Oscillator and Decade Scaler

The General Radio LR-3 Combined Hetereodyne Frequency Meter and Crystal-Controlled Calibrator, hereafter referred to as a crystal oscillator, has a rated output specification of 100 KC$\pm$ one cycle at 50°C. The crystal oscillator was operated, however, in its 10,000 counts-per-second mode. This output was routed to a Technical Associates DS-5B Decade Scaler which has a rated resolving time of 5 microseconds. The scaler was modified so that pulses from its fourth decade (1 cps) were tapped. Furthermore, they were appropriately shaped for input to the chart recorder. Thus a highly accurate series of one-second pulses was generated. The crystal oscillator and this scaler will hereafter be referred to as the crystal oscillator-scaler linkup.

Chart Recorder

A Brush Mark II recorder, hereafter referred to as chart recorder, was used during coincidence runs to record coincidence count blips and time marks on chart paper which has longitudinal and transverse grid lines. The chart recorder has two primary channels, two constant-amplitude event-marking channels, and push-button-controlled chart speeds (1, 5, 25, and 125 mm/sec). For coincidence data runs, one channel (count channel, Figure 5) received pulses from the coincidence module-scaler linkup and the other channel (time channel, Figure 5) received pulses from the crystal oscillator-scaler linkup. By using the timing marks of the time channel as the reference for time, little dependence was placed on the accuracy of the chart speed for coincidence count rate data. In all runs the chart recorder was operated at 25 mm/second.

Mercury Pulser

Pulses from the Hamner NP-10 Mercury Switch Pulse Generator were used to simulate the output pulse characteristics of the nuclear radiation detectors for purposes of testing and calibrating most components, and combinations thereof, of the entire counting system. The mercury pulser has a repetition rate of 60 cps (or line frequency) with selection of variable rise and decay times for its pulses. It has helipot-controlled pulse heights whose height stability is rated at 0.002% per volt change in line voltage. The mercury pulser was used, in conjunction with a multichannel pulse height analyzer, to calibrate the E and

ΔE dial settings of the single channel pulse height analyzers for coincidence runs. When set at a rise time of 250 nanoseconds and a decay time of 100 microseconds, the pulser output resembled Cs-137 pulses from either detector number one or detector number two amplifier channels.

Multichannel Analyzer

The Nuclear Data ND-180 FM Analysis System, together with some of its Readout Accessories, was employed during this experiment. It was used for obtaining pulse height (energy) spectra, for calibration, and for gain-shift monitoring. Of the 512 available channels, 256 were used. Each spectrum output consists of a typed page of 256 numbers, in channel order, where each number represents the number of counts recorded in its respective channel. The system has a rated dead time of 5 microseconds and stability of less than one channel drift per day.

4. Calibration.

Amplifier Gains

A circuit arrangement similar to Figure 4, except that the mercury pulser was replaced by a Cs-137 source, was used to adjust the gain setting on both amplifiers. The E dial of each single channel analyzer was set at eight volts (ΔE at .25 volts) and then the gain of each amplifier was adjusted until the Cs-137-.662 mev photopeak appeared at this window setting. The geometry of both detectors and the Cs-137 source holder was essentially permanent (Figure 6) during

the entire experimentation period. Any subsequent drifts in amplifier gain would shift the Cesium peak relative to the above pulse height analyzer settings. Therefore, this calibration provided a method for monitoring gain-shifts in the circuits of either detector number one or detector number two.

Resolving Time of Coincidence Module

Two independent Cs-137 sources were used to ascertain the resolving time of the coincidence module whose helipot dial (0.00 to 10.00) was set at 5.00. Detector number one and detector number two were separated by approximately two feet and sufficient shielding was used to prevent detector number one from receiving any pulses from the source being counted by detector number two and vice versa. Using the relation: (chance coincidence count rate) = 2 (resolving time) (singles count rate of detector number one) (singles count rate of detector number two), the resolving time, t, was found to be 75 nanoseconds at a helipot setting of 5.00. This value represents the average of three measurements and has a statistical error of 1.1%.

Energy Calibration of Multichannel Analyzer

An approximate energy calibration for the first 256 channels of the multichannel analyzer was a prerequisite for future uses. This multichannel pulse height analyzer was to be used in a direct manner to calibrate the pulse height intervals of both single channel pulse height analyzers.

The mercury pulser was used to adjust the multichannel analyzer so that its channel one corresponded to zero pulse height. Subsequent responses of this analyzer were found to be very linear with respect to the dial setting of the linear mercury pulser. However, the Cs-137 .032 mev (X-ray peak) and .662 mev (gamma photopeak) pulses, when routed over either detector channel (Figure 2), did not quite give an energy calibration in which zero energy corresponded to channel one. The slight shift reflected the well-known fact that scintillator counters exhibit some degree of nonlinearity at low energies. However, the assumption that the energy calibration of the multichannel analyzer was linear up to .662 mev with zero energy in channel one was deemed a reasonable approximation.

Window of Single Channel Analyzer Number One

With the activated Hafnium sample in the counting position (Figure 2), the output of amplifier number one was fed directly into the multichannel analyzer. The resulting pulse height spectrum was very similar to that of Figure 3. It was decided to have the coincidence input of single channel analyzer number one consist of all pulses greater than .15 mev (channel 28 of multichannel analyzer). This corresponded to a maximum singles counting rate of 60,000 counts per second which was desirably high for coincidence count rate purposes, but still sufficiently low to give a reasonable chance count rate. Additionally, this decision meant that the undesirable .06 mev reflection peak would not

be included. Lastly, this lower threshold of .15 mev was sufficiently below the .22 mev photopeak to minimize any effects due to possible gain-shifts in detector number one.

The next procedure was to adjust the E dial of single channel analyzer number one (integral mode) to correspond to the above chosen pulse height of .15 mev. This was done by putting mercury pulses into detector number one and then routing the output of amplifier number one directly into the multichannel analyzer (Figure 4). The height of the mercury pulses was varied until the multichannel analyzer indicated pulses in channel 28 (.15 mev). At this point the output of amplifier number one was rerouted directly into single channel analyzer number one (Figure 4), whose E dial was set at zero. Next the E dial setting was adjusted until the output pulses of this single channel analyzer just disappeared, as indicated by the scaler.

Window of Single Channel Analyzer Number Two

The Hafnium pulse height spectrum from amplifier number two was obtained, as described above, using the circuitry illustrated by Figure 2. The pulse height interval on this spectrum (Figure 3) between .29+ mev (channel 55 of multichannel analyzer) and .48+ mev (channel 89) was chosen as the coincidence input from single channel analyzer number two. This pulse height interval would include the .326 mev and .427 mev gammas of Hf-178m (Figure 1) but hopefully would exclude all pulses from the Hf-179m (Figure 1).

The E and the ΔE dials of single channel analyzer number two (differential mode) were adjusted to prevent all pulses of energies (pulse heights) below .29+ mev or above .48+ mev from entering the coincidence module. These adjustments were accomplished using a calibration technique which was a minor extension of the method employed for adjusting the E dial of single channel analyzer number one (integral mode). The calibration technique required the use of the mercury pulser and the multichannel analyzer.

5. Procedure.

Systems Checkout

A dummy projectile was used to test the proper functioning of the penumatic transfer system. Both channels of the chart recorder were operationally tested using either 60 cps or one-cps inputs. Using an electric timer, a one-minute run of the crystal oscillator-scaler link-up and the chart recorder was conducted to detect any malfunctions in the timing system. A circuit arrangement, similar to Figure 4 except that the mercury pulser was replaced by a Cs-137 source, was used for a gain-shift check of each detector channel. The ΔE dial of each single channel analyzer was set at .25 volts. Then the E dial of each single channel analyzer, in turn, was varied until the Cesium .662 mev photopeak was located, as indicated by the scaler. The resulting E dial setting of each analyzer was compared to the previously obtained E dial setting of eight volts (See Amplifier Gains, Section 4.) for the

Cesium photopeak. Any drift from eight volts would indicate a long-term gain-shift in that particular detector channel.

Preliminary Pulse Height Spectra

Prior to actual data runs the multichannel analyzer was used to obtain pulse height spectra of both activated Hafnium and Cs-137 separately. These spectra, which were recorded on the typed output of the multichannel analyzer, provided a reference for future measurements of gain-shift.

Coincidence Data Collection

Backgrounds of both the coincidence count rate and the detector number one singles count rate were made with the AGN-201* reactor stabilized at eight watts. By actuating the pneumatic transfer controls the sample was irradiated for 10.2 seconds (one run was 10.4). By the time of arrival of the irradiated Hafnium sample at the detectors' position, the chart recorder was already in operation. It was operating at 25 mm/second and recording timing marks from the crystal oscillator-scaler linkup (Figure 5). The initial spacing between recorded coincidence count (scaled) blips was approximately 3/16 inch. As the spacing between these blips on the chart paper increased to

*The AGN-201 is a graphite and polyethylene-moderated research reactor capable of producing a neutron flux up to 4×10^4 n/cm^2-sec at the AEC license limit of 1000 watts. [2]

about 3/4-inch, the scaling factor for each coincidence blip was reduced. This was done by turning the scale selection dial to the next lower power of ten. This changing of scale factor continued until each blip represented only ten coincidence counts. When the distance between successive coincidence count blips on the recorder output approached about six inches the recorder was stopped. This constituted one coincidence-data-collection run; the output consisted of chart paper inked with scaled coincidence blips, one-second timing marks, and event marks on the margin. The event marks were electronically triggered by the switches which moved the projectile into the core or fired it back to the counting position. They provided measures of how long the firing switch was actuated and of the time length of irradiation of the sample.

No subsequent run was conducted until the detector number one singles background returned to essentially its original value. The typical time spacing between each two consecutive runs was of the order of five minutes.

Interspersed among the several data collection runs were checks for medium-term gain-shift. These consisted of obtaining the pulse height spectra of both the Hafnium sample and Cs-137. Using the mercury pulser, the E setting of analyzer number one and the E, ΔE settings of analyzer number two were checked against the previously determined multichannel analyzer addresses. All checks revealed no measureable spectral shifts nor shifts of the single channel window

settings relative to their reference addresses in the multichannel analyzer.

Coincidence Versus Singles Data

In order to obtain data for making corrections for chance counting rate, two special runs were conducted. The crystal oscillator-scaler linkup was replaced (Figure 5) in turn by the output of single channel analyzer number one and single channel analyzer number two. With the above modifications, coincidence-versus-singles data runs were conducted as described above,for the coincidence-versus-time data runs. Thus the recorder output did not include one-second timing marks; however, the nominal chart speed of 25 mm/second was used as a time reference, sufficiently accurate for applying the chance counting rate corrections.

6. Data reduction and analysis.

Coincidence Counts versus Time

On the outer edge of the recorder chart paper (See Chart Recorder, Section 3.) there were event marks, the distance between which was a reasonable measure of irradiation time. Additionally, there were one-second timing marks along one side and scaled (10^4, 10^3, 10^2, or 10) coincidence count blips along the other side. All counting intervals began and ended on count blips. The displacements in centimeters (measured to the nearest hundredth of a centimeter with a quality

engineer's scale) of these count blips from nearby time marks were measured. These measurements provided sufficient information to compute the beginning time of a counting interval and the time duration of that counting interval. The number of counts in the interval was given by the product of the integral number of count blips and the count-blip scale factor. The time, counts, and distance data from these charts provided an input for the data reduction computer program, REDUCTION.

Coincidence Counts versus Singles Counts

On one side of the chart output there were scaled coincidence count blips and on the other side there were scaled count blips from the auxiliary scaler connected to the single channel analyzer of detector number one (or detector number two). Since there were no timing marks, the nominal chart speed of 25 mm/second was used to determine the time scale. Precise measurement of the chart speed indicated that it is essentially constant and within 1% of 25 mm/second. As described above, the time, counts, and chart distance data were extracted from the chart of coincidence-versus-analyzer number one singles. This data and also that of coincidence counts-versus-analyzer number two singles were processed, in turn, by the REDUCTION computer program. This program, briefly, takes this input data and gives as primary output for each counting interval:

T(I). . . . . .beginning time

DT(I). . . . . .time duration

C(I). . . . . .counts

TMID(I). . . . . .time at midpoint

CR(I). . . . . .count rate at midpoint

The CR of coincidence and singles number one were plotted against a common time reference. A similar plot was made on a separate graph involving singles number two. Since both initial (extrapolated to time zero) coincidence count rates (2770 cps) were identical, the two singles rates were assumed to be normalized and were plotted versus the same time scale on the same graph. Using the above graph and $2t = 150$ nanoseconds, the product $2t\, \dot{N}_1\, \dot{N}_2$ (where the last two terms are the two singles' rates) was computed for each one-second point and plotted. This then gave chance count rate ($\dot{N}_{ch}$) versus TMID for an initial coincidence count rate of 2770 cps.

Chance Counting Rate Correction

From the REDUCTION computer program output for each regular (coincidence counts versus time) data run, a plot was made of CR versus TMID and extrapolated to time zero for comparison with 2770 cps. The factor

$$F = \left(\frac{\text{zero intercept CR}}{2770} \right)^2$$

for each run was computed. For each counting interval of a run, a chance count correction to C(I) was computed. TMID(I) was used to

enter the chance rate graph ($\dot{N}_{ch}$ versus TMID) and then the product

$$F \times (\text{chance rate}) \times DT(I)$$

was computed. This product was subtracted from C(I) to yield the corrected counts in the I'th counting interval, C'(I). The processed data, T(I), DT(I), and C'(I) formed the essential inputs for the data analysis computer program, FRANTIC.

FRANTIC Computer Program

The FRANTIC Program for Analysis of Exponential Growth and Decay Curves [5] can accomodate up to 400 data points and analyze a single composite curve consisting of a sum of no more than ten components, each of the form $A_i e^{\lambda_i t}$. It processes raw counting data and fits to these, by least squares analysis techniques, a sum of exponentials of the above mentioned form. The A_i is the counting rate at time zero; the λ_i is the decay constant.

The salient features of its input include:

Number of components

Holding parameters (A and/or λ) fixed (optional)

Statistical weighting

Dead time corrections

Background (optional)

Uncertainty in timing interval

The salient features of its output include:

Number of iterations

Number of points deviating more than 2 sigma (sigma is an estimator of the standard deviation)

A histogram of such deviations

The weighted variance of fit (ideally 1)

Chi square and degrees of freedom (ideally the former ÷ latter = 1)

Activity at time zero in counts per unit time and its sigma

Half-life and its sigma

7. Error Analysis.

Sensitivity of FRANTIC Computer Program to Number of Input Data Points

The number of data points which can be extracted from a given chart recorder output (scaled count blips and time marks) is a parameter under the control of the experimenter. For example, the entire interval of time on this chart paper between the first and last count blips can be divided into a maximum number of sub-intervals equal to the total number of blips (less one). Each sub-interval begins on a count blip, can include several blips, ends on a count blip, and constitutes a data point.

An earlier study was made to determine the effect of the number of data points on the FRANTIC-computed half-life of two radioactive isotopes whose half-lives were of the order of five seconds. For each isotope a single 24 data point set was available. By selecting progressively longer sub-intervals of time, four additional sets (12, 8, 6,

and 4 points) were made. All five sets, in turn, were analyzed by the FRANTIC program and the resulting half-lives and their sigmas (estimates of the standard deviations) were studied. Both the half-lives and their sigmas differed only negligibly for the five cases considered. There was no meaningful pattern of variation of either as functions of the number of input points. Returning the present work, the number of sub-intervals extracted from each Hafnium chart output resulted in counting-time intervals of approximately two seconds for the first three or four points and approximately four seconds for the remaining data points. These counting-time intervals of two and four seconds were approximately equal to those of the previously mentioned 24 and 12 point sets respectively. Therefore it is believed that no appreciable errors were introduced by choosing the number of data points in the manner indicated above.

Static Equilibrium of the Projectile

During actuation of the firing switch, air at high pressure propelled the light weight projectile to the counting position. On arrival, the conical nose of the projectile collided with a female conical bronze fitting which was fitted with a high pressure air-bleed-off valve. The escape of this air was accompanied by a squealing type noise which lasted approximately as long as the firing switch was actuated (approximately one second for all runs except one). During this short period of time the projectile is believed to be oscillating into and out of

the desired final counting position. During the "out" portion of the oscillation, less counts will be detected than for the "in" position; such depression of initial counting rate lengthens the computed half-life. It was essential to determine from the first few data points of each run that time where oscillations had ceased and the projectile was at rest.

If the above assumptions are valid, then by removing the first, second, third, , , , data points in turn, the resulting computed half-lives should decrease to a statistical plateau. This point stripping was performed on the inputs for the FRANTIC program and the results were as follows:

Computed Half-life in Seconds

Data Run	Number of points stripped		
	0	1	2
1	3.97	3.92	3.92
2	4.01	3.99	4.00
3*	4.04	4.03	3.98
4	3.89	3.83	3.82
5	3.98	3.96	3.95
6	3.99	3.95	3.95
7	3.99	3.96	3.92
Average	3.98	3.95	3.94

In all runs the first three data points each had counting intervals of approximately two seconds. The above tabular results support the

*Firing switch was actuated for about two seconds.

conclusion that by the time data was being recorded for the second point, the projectile was at rest, provided the firing switch was actuated for no more than one second.

Effect of Background Uncertainty on Half-life

To ascertain the effect of background on the FRANTIC-computed half-life of Hf-178m, six different backgrounds for the seventh data run (centered on the computer determined background) were assumed. The percentage errors in half-life at the extremes (which were 32% and 160% of the computer determined background) were 0.1% and 0.22% respectively. Fifteen coincidence counts recorded during sixty seconds was a typical measured background; the statistical uncertainty in the corresponding count rate is only 24%. Thus it was concluded that the error in half-life attributed to background uncertainty was .1% or less.

Effect of Resolving Time Uncertainty on Half-life

To ascertain the effect of resolving time on the FRANTIC-computed half-life of Hf-178m, two extreme deviations from the previously calculated resolving time (75 nanoseconds) were used. The deviations used were 56 and 94 nanoseconds which were 75% and 125% respectively of the 75 nanoseconds. They resulted in half-lives which differed from the original (based on 75 nanoseconds) by percentage errors of .025% and .18% respectively. Thus, since the 75 nanosecond resolving time was determined with a precision of 1.1%, it was concluded that the

effect of resolving time uncertainty on the half-life of Hf-178m was negligible.

Effect of 18-second Hf-179m Component on Half-life

One coincidence data run was selected for analysis of possible effect of counts due to Hf-179m decay. From the REDUCTION computer program output, CR versus TMID (See Section 6.) was plotted on semi-log graph paper. From all points on this curve the statistically minimum background was subtracted. The resulting points were strictly linear from 2500 cps to below 1 cps. If any Hf-179m (18-second half-life) activity were present, then the above plot indicated that its initial activity was less than one count per second. Such a low initial activity would mean that the Hf-179m contribution to each point on the decay curve would be smaller than the statistical fluctuation of that point.

A single chance coincidence count can result from the gamma emissions of two or more Hf-179m nuclei. The time distribution of such chance counts does not, in general, follow the Poisson distribution whose time parameter corresponds to the 18-second half-life of Hf-179m. Hence the correction for counts under these circumstances was included in the chance count rate correction (See Chance Counting Rate Correction, Section 6.).

If a coincidence count could be triggered by the gammas of a single Hf-179m nucleus then the occurrence of such counts would

follow a Poisson distribution. Furthermore, such a distribution of coincidence counts would constitute an 18-second decay component. As a consequence, the decay curve resulting from all coincidence counts would be the sum of two component decay curves (Hf-179m plus Hf-178m). Then, in order to obtain the Hf-178m decay curve, it would be necessary to subtract the Hf-179m component from the composite decay curve. However, in order to get a coincidence count from a single Hf-179m nucleus, it is necessary that both the .215 mev and the .160 mev gammas (Figure 1), or gammas of lesser energies arising from them, arrive at detector number two extremely closely in time. The detector, unable to distinguish them, passes on a single pulse whose energy (pulse height) will be close to their algebraic sum. Such an event is referred to as a pulse pile-up. Since there are no remaining gammas from that decaying Hf-179m nucleus (Figure 1) to go to detector number one, then no coincidence count can be generated. Therefore, it is concluded that the decay curve derived from all coincidence counts does not include a Hf-179m component.

Gain-shift

During the course of this experiment, only checks for long-term and medium-term gain-shifts of the detector channel were conducted. These checks were made using a Cs-137 source (See Calibration, Section 4. and Systems Checkout and Preliminary Pulse Height Spectra, Section 5.); they were also made using the activated Hafnium sample

(See Preliminary Pulse Height Spectra, and Coincidence Data Collection, Section 5.). All checks revealed that no spectral shifts occurred. Furthermore, observations showed that there were no shifts of the windows of the single channel analyzers relative to their reference addresses in the multichannel analyzer.

The number two detector was used by Button et al [1] in 1965 for half-life measurements. At that time the detector was modified so that the detector gain could be monitored while decay data was being collected. They did monitor the detector gain while the activated Hafnium sample was irradiating the detector. This experiment indicated that any variations in gain which did occur in the detector were indistinguishable from statistical fluctuations in the measurement of that gain. One run, which they made using a different element, indicated the possibility of a slight shift of gain just outside the uncertainty of the gain measurement. It was assumed that the overall change in gain was .25% which was further assumed to vary linearly over the period of decay. The FRANTIC computed half-life, based on the above assumptions, deviated .6% from the half-life computed for no gain-shift.

Additional measurements were later carried out by Rodeback, G. W. (private communication from Rodeback) with the above equipment wherein he used several different low energy gamma sources (including Hafnium). These measurements indicated no detectable gain-shift in detector number two during the time of decay of these sources. His estimate of maximum gain-shift during any of the above measurements was about .1% which would correspond to about .2% in half-life.

Based on the performance of both detectors, noted during this experiment, and the findings of Rodeback and Button et al, it is concluded that the effects of gain-shifts on half-life determinations for Hf-178m were small. It is further concluded that .4% represents a reasonable estimate of the maximum error in half-life due to possible gain-shifts in the circuits of detector number one and/or detector number two.

8. Acknowledgements.

The author wishes to express his appreciation to his thesis advisor, Professor G. W. Rodeback, whose invaluable patience and astute guidance greatly contributed to the execution of this work. Furthermore, the author also desires to acknowledge the assistance of Mr. H. L. McFarland who skillfully operated the reactor and kept all associated electronics functioning.

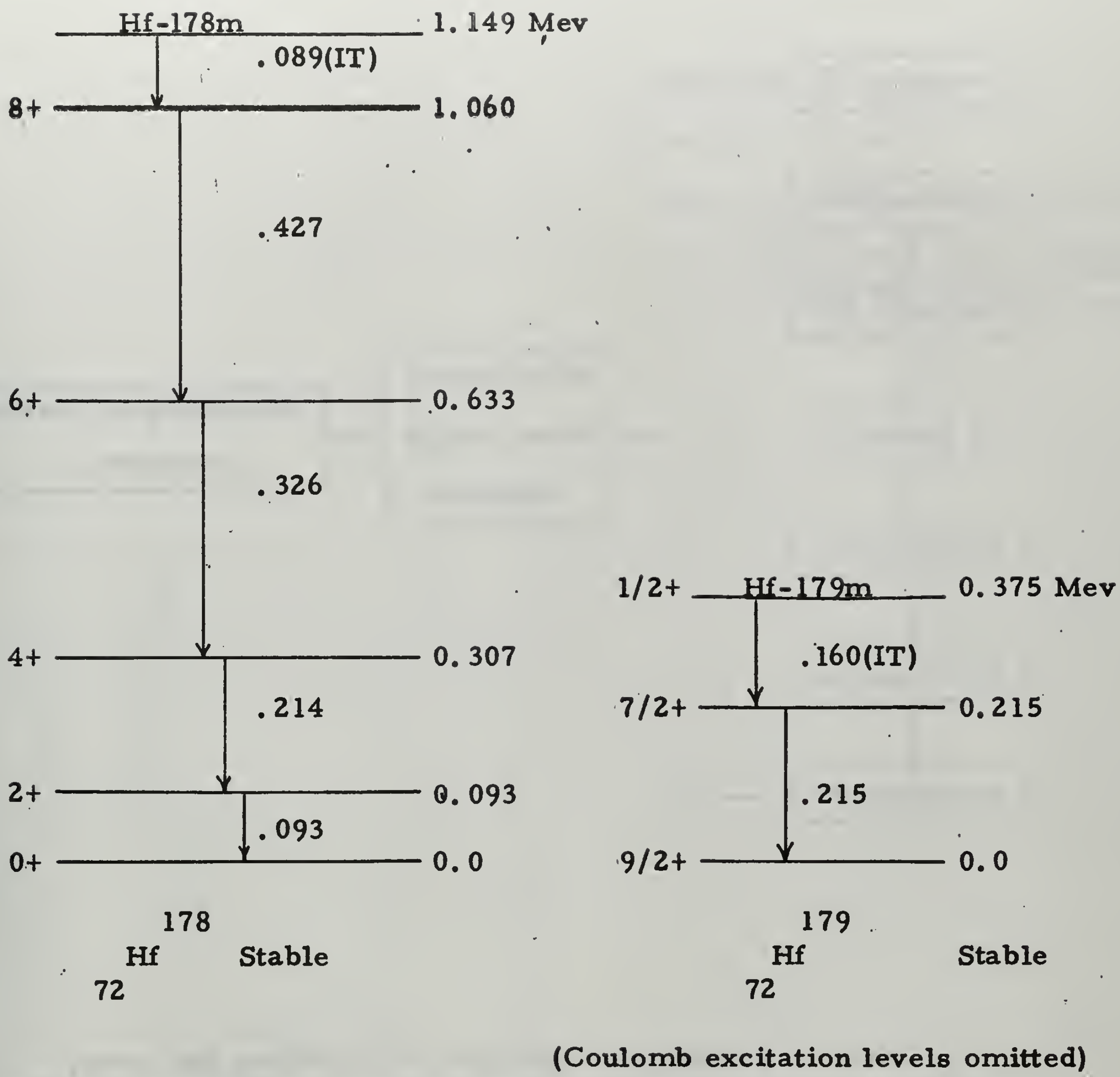

Figure 1. Gamma decay schemes[1] of Hf-178m and Hf-179m

[1]Strominger, D., Hollander, J. M., and Seaborg, G. T., Table of Isotopes.[7]

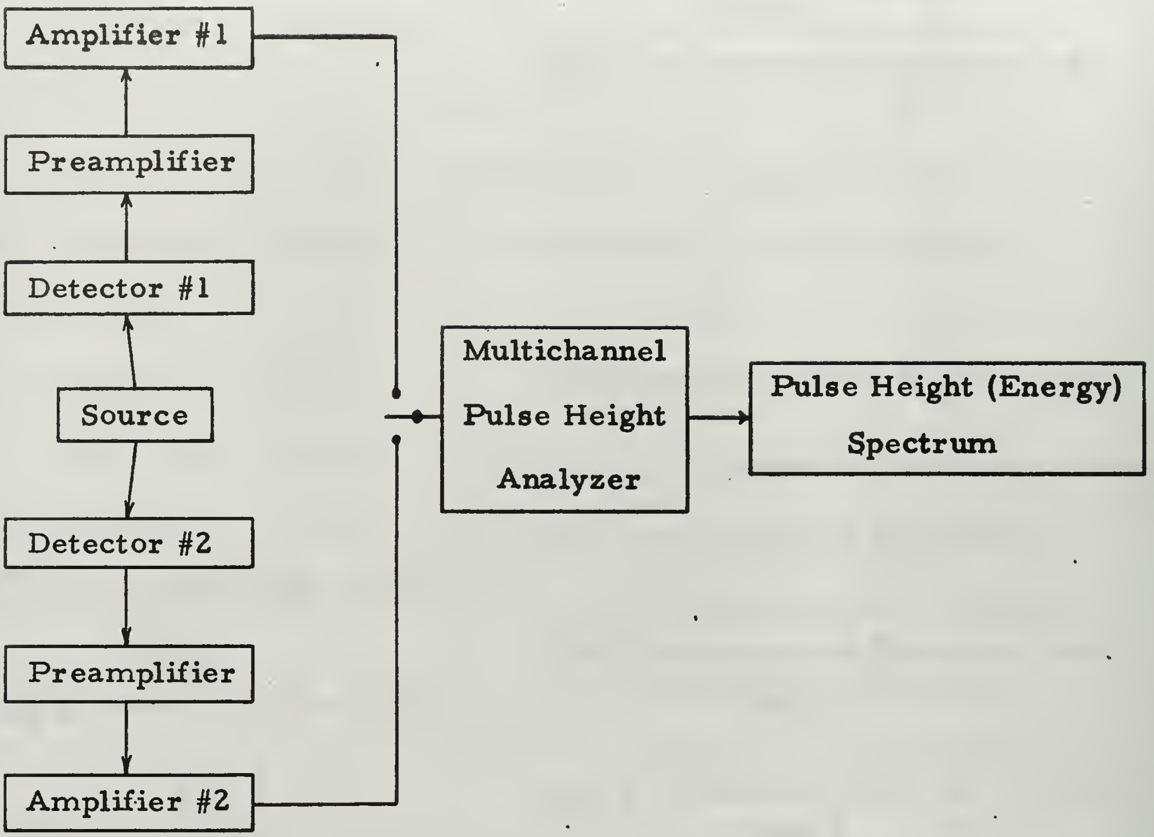

Figure 2. Schematic of circuitry for obtaining the energy spectrum of either Cs-137 or the mixture of Hf-178m and Hf-179m for either Detector #1 or Detector #2.

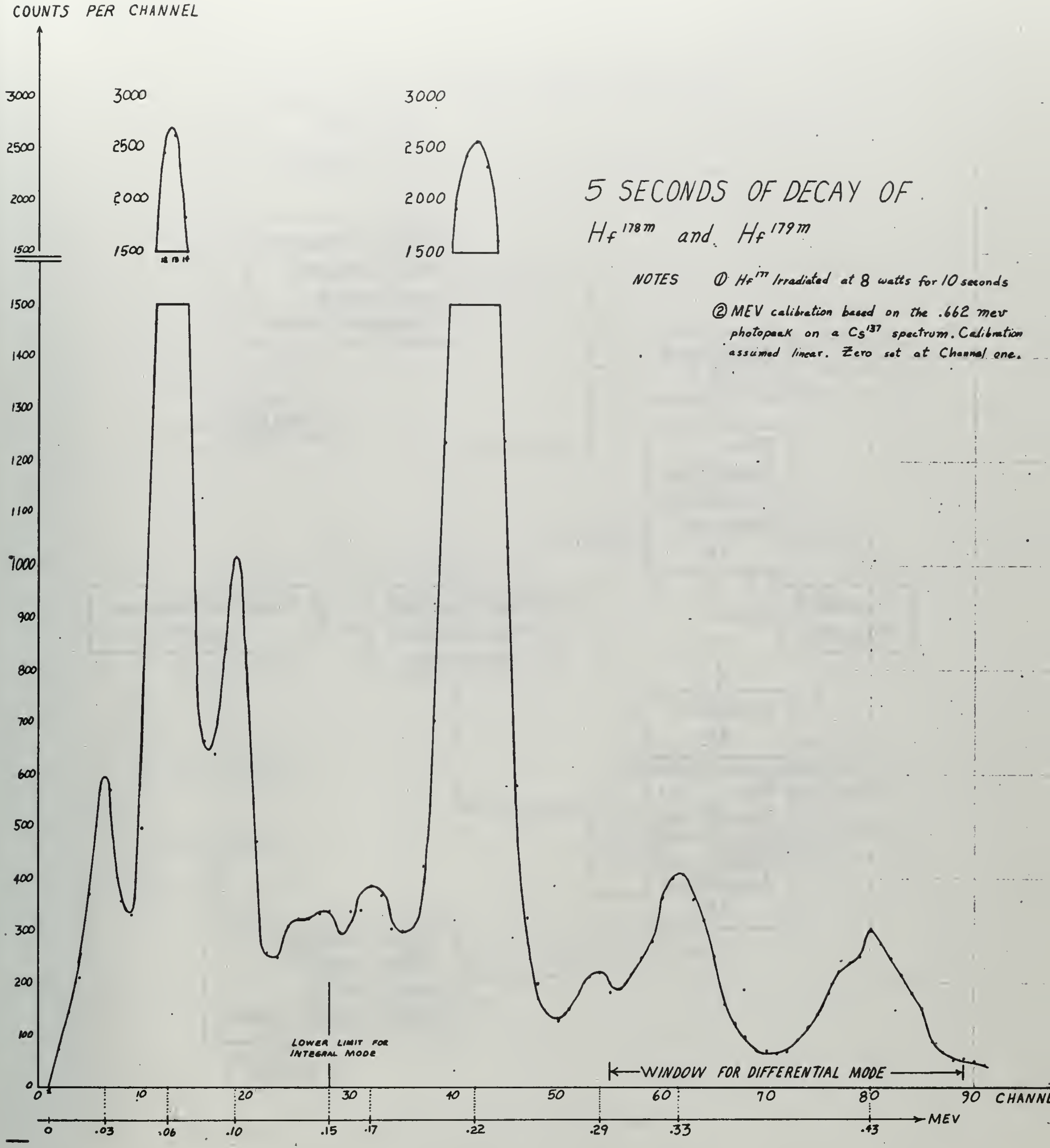

Figure 3. Pulse height (energy) spectrum of mixture of Hf-178m and Hf-179m

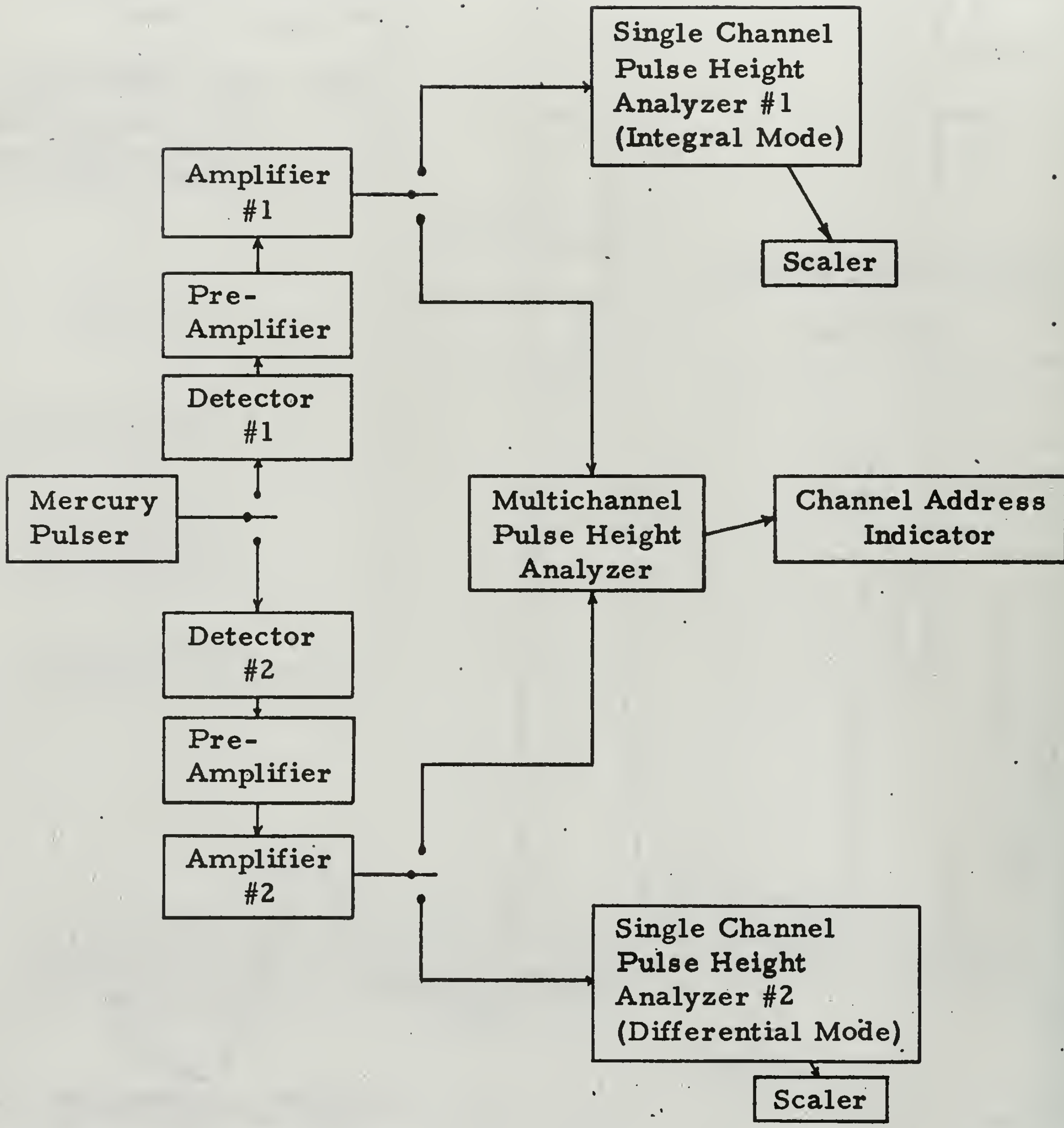

Figure 4. Schematic of circuitry used to calibrate the E dial setting for Pulse Height Analyzer #1 and both the E and ΔE settings for Pulse Height Analyzer #2.

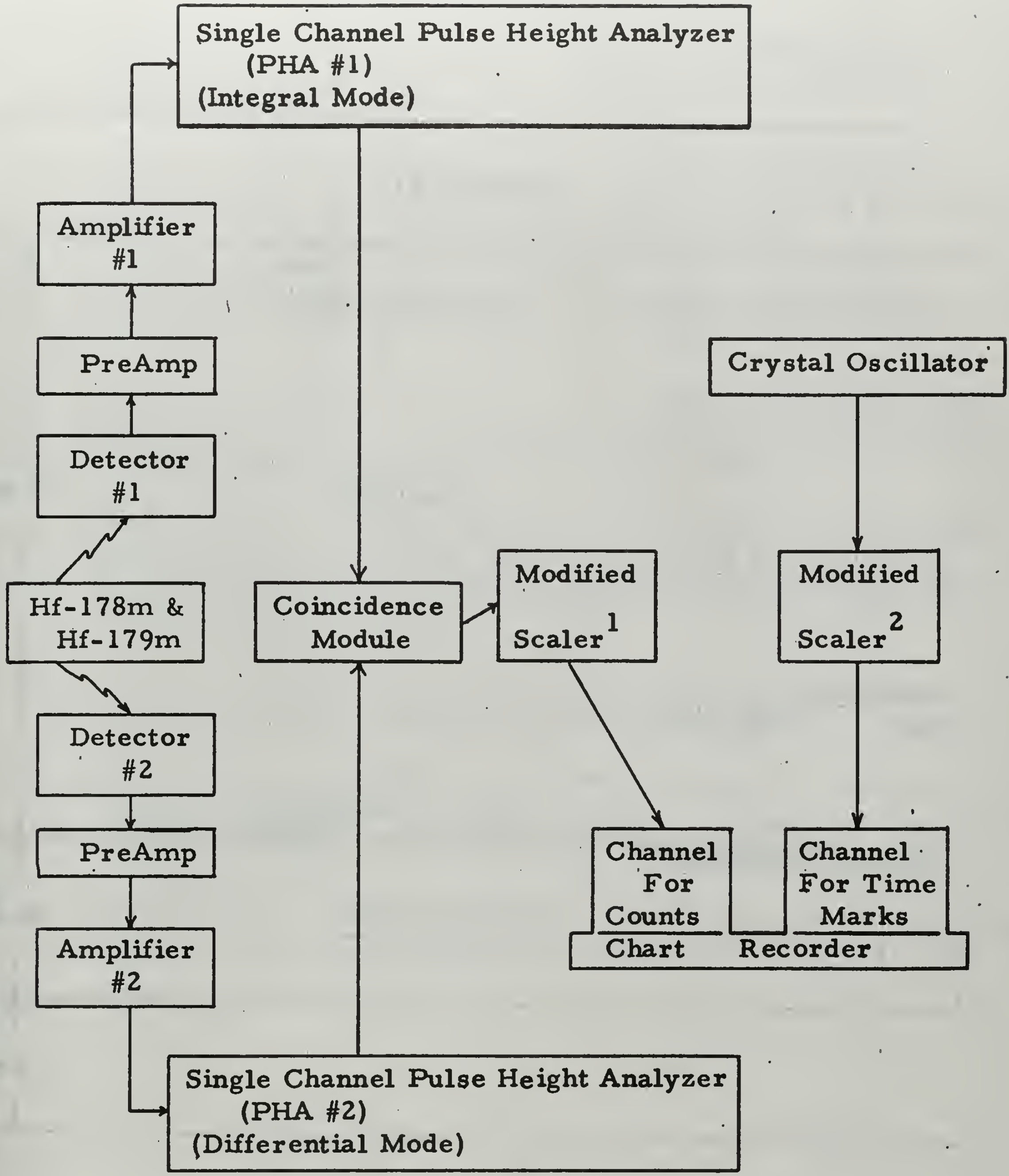

Figure 5. Coincidence data collection

[1]Variable scaled output (10, 10^2, 10^3, 10^4).
[2]Scaled output of 10^4.

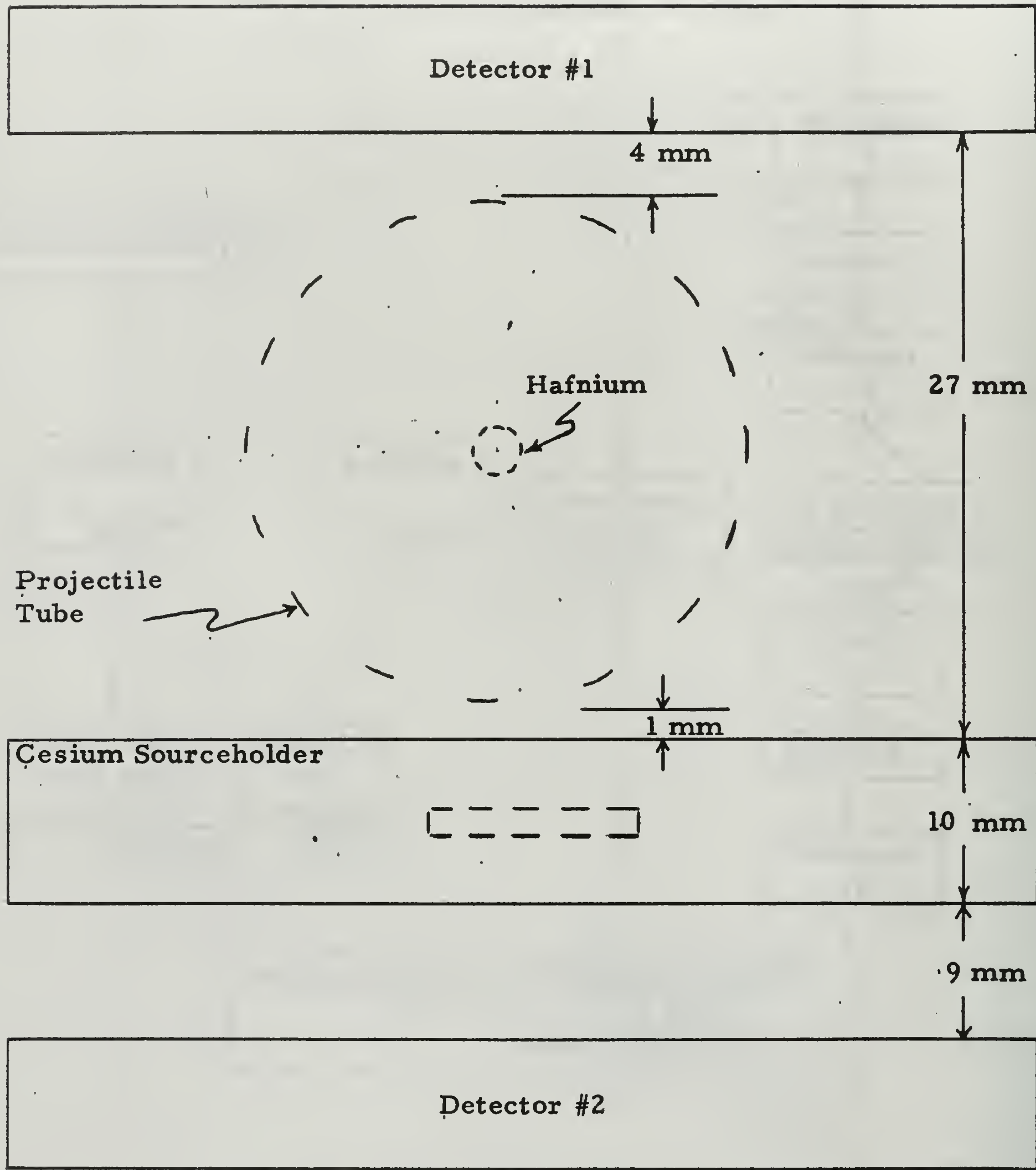

Figure 6. Schematic of counting geometry.

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